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(54) Name of the invention: Manufacturing Method for the Preparation of Pressure Sensitive Adhesive Tape

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(54) Manufacturing Method for the Preparation of Pressure Sensitive Adhesive Tape

Detailed Explanation of the Invention

The present invention is an invention about the method for the manufacturing of an excellent adhesive tape where there is no separation of the adhesive agent from the tape substrate material. And especially, the present invention is an invention about a method that is appropriate for the manufacturing of an adhesive tape which uses as its substrate material a synthetic resin film that has relatively poor adhesive properties, like polyethylene, polypropylene, tetrafluoroethylene etc.

In the past, in the case when polyethylene, polypropylene, tetrafluoroethylene etc., synthetic resins were used as the tape substrate material, the above synthetic resin films were materials that have excellent chemical and electrical properties, however, it has been difficult to manufacture good adhesive tapes because of the fact that their adhesive properties relative to the adhesive agent, are poor. However, until now, different studies (research) have been conducted in order to strengthen the adhesion of the synthetic resin films possessing poor adhesive properties and the adhesive agent. Among these, as it has been reported in the description of the Japanese Patent Report Number Showa 40-24788, it is known that by the method where an adhesive agent to which a reduction sensibilization agent has been admixed, has been coated on the surface of the tape substrate material, and after that, from the back surface of the tape it is irradiated using a visible or ultraviolet light, and by that it is possible to strengthen the adhesion between the tape substrate material and the adhesive agent. However, that notwithstanding, in the case of the method where this way by irradiation of visible or ultraviolet light at the interface between the tape substrate material and the adhesive agent a crosslinking reaction between the two materials occurs, and based on that the anchoring strength is increased, there is the drawback that this method is limited to the cases where as the

tape material a material is used that is relatively thin and not only that, but also it is a material that has good transparency properties. And also, in the case of the adhesive tape obtained this way, for example, even when a radiation beam sensibilization agent has been compounded into the adhesive agent and the crosslinking reaction is continuously conducted, it has been difficult to completely finish such crosslinking reaction and because of that there is the strong trend that with the passing of days, gradually the crosslinking reaction proceeds and the adhesive properties are lost.

Especially, also, even according to this method, at the time when polyethylene, polypropylene, tetrafluoroethylene etc., resins are used as the tape substrate material, it has not been possible to obtain sufficient adhesive strength between the tape substrate material and the adhesive agent.

However, the present invention is an invention according to which a crosslinking reaction which is extremely difficult to control, such as the one described here above, is not used, and it is an invention where by the irradiation with ionization radiation beam the vinyl type monomer, which has been added in the adhesive agent, is polymerized and at the same time it is also graft polymerized onto the tape substrate material and together with that onto the adhesive agent component, and by that the anchoring force of the adhesive agent relative to the substrate material is extremely improved.

Namely, the present invention is an invention about a method for the manufacturing of pressure sensitive adhesive tape, characterized by the fact that an adhesive agent containing vinyl type monomer material, is provided on the surface of the tape substrate material, that is formed from synthetic resin film, and then, towards that an ionization radiation beam is irradiated.

Regarding the term "adhesive agent" used according to the present invention, it has the meaning of all adhesive agents usually used for adhesive tapes. Regarding the usually used adhesive agents, they are formed as adhesion imparting agent is added to rubber or synthetic resin, and they are materials where, especially, depending on the requirements, it is possible to add plasticising agents, anti-oxidation agent, filler agent, pigmenting material etc., and naturally all of these can be used according to the present invention. Also it is a good option if they are polymer materials that have been obtained as vinyl type monomer material has been appropriately synthesized.

As the rubber material, it is possible to use natural rubber, styrene – butadiene rubber, nitrile rubber, butyl rubber, neoprene rubber, synthetic – natural rubber, etc., and also, as the synthetic resins, it is possible to use polycondensed materials that have been obtained by the appropriate synthesis of thermoplastic type resin materials like vinyl chloride resin, vinyl acetate resin, vinyl chloride – vinyl acetate copolymer etc., and of vinyl type monomer material etc.

Also, as the adhesion imparting agent it is possible to use terpenic type resins, which use α and β pinene as their main component, rosine type resins, which use abietic acid as their main component and modified rosine materials obtained by the hydrogenation or esterification of rosine type resins, hydrocarbon type resins obtained from olefin type materials, etc.

As the adhesive agent composition material that is used according to the present invention, a material is used that is obtained as to such rubber type or synthetic resin type adhesive agent vinyl type monomer material is admixed and incorporated.

As the vinyl type monomer material it is possible to use material from gas to solid phase materials, however, materials like, styrene and styrene derivative materials, methacrylic acid, acrylic acid, acrylamides, vinyl acetate, unsaturated dibasic acid derivative materials like esters of the maleic acid, esters of the acrylic acid, methacrylic acid esters, vinyl esters and vinyl ethers, etc., which are liquid state or solid state materials under normal temperature are preferred from the point of view of ease of the procedures of mixing these materials into the adhesive agent.

Especially, butyl acrylate, vinyl acetate, methyl methacrylate, are preferred.

If the material used as the tape substrate material is a synthetic resin film, it is very effective because it is an appropriate material, and although there are no specific limitations, the present invention is especially effective at the time when polyethylene, polypropylene, tetrafluoroethylene, etc., synthetic resin films, for which the adhesion between the substrate material and the adhesive agent has been difficult to achieve according to the previous technology, are used as the tape substrate material. After that, the manufacturing method for the preparation of the adhesive tape according to the present invention will be described schematically.

First, the natural rubber or synthetic rubber material, which forms the main component of the adhesive agent, was mixed by using a roll or a Bambarry mixer, and after that it was appropriately cut and this was introduced in a dissolution tank where an organic solvent agent had been added in advance. Especially, to that the appropriate amounts of adhesion imparting agent, and vinyl monomer material, and also, depending on the requirements, plasticizing agent, and anti-ageing agent, were sequentially mixed in and introduced, and as this was heated it was stirred and dissolved. The obtained by this adhesive agent composition material was coated on one or both surfaces of the tape substrate material by using roll coater, knife coater, etc., coaters etc., so that the thickness becomes constant. After that, from one surface of this tape ionization radiation beam was irradiated and after that this was passed through a drying device. At the time when this material is passing through the drying device, the solvent agent in the adhesive agent and the unreacted vinyl type monomer material, were almost completely volatilized.

Moreover, in the case according to the above described method, the solution of the adhesive agent composition material was coated and after irradiation by using ionization radiation beam, it was dried. However, also in the case when the solution of the adhesive agent composition material is coated and dried and the solvent agent inside the adhesive agent and the excess vinyl type monomer material, are volatilized, and then after that it is irradiated by using ionization radiation beam, the same results are obtained.

Especially, in the case of the present invention, modifications such as those described here below are also possible. Namely, natural rubber or synthetic rubber material is mixed and kneaded by using double roll or Bumbarry mixer, and together with that to that the appropriate amount of adhesion imparting agent, plasticising agent, filler agent, anti-ageing agent, and vinyl type monomer material, are sequentially admixed and incorporated, and by that a solid phase adhesive agent composition material is obtained.

The obtained by this adhesive agent composition material is coated, by using a calender etc., on one surface side of the tape substrate material, so that it becomes a layer with a constant thickness. After that, from one surface of this tape ionization radiation beam is irradiated, and after that this was cut to constant length as it is in a sheet shape, or it is wound to the appropriate length and after that it is cut at constant length, and roll shape manufactured product is obtained.

Especially, in the case of the present invention, it is also a good option if before conducting the polymerization of the vinyl type monomer material by the irradiation with the ionization radiation beam, it is partially polymerized by employing a catalyst, and then, after that, it is polymerized by ionization radiation beam.

According to the present invention, it is considered that by coating a vinyl type monomer material containing adhesive agent composition on one surface side of the tape substrate material, and irradiating towards that ionization radiation beam, the vinyl type monomer material that has been incorporated into the adhesive agent, is itself polymerized, and at the same time, one part of the vinyl type monomer material is also graft polymerized onto the tape substrate material surface and together with that onto the adhesive agent component, and by that the anchoring force of the adhesive agent relative to the tape substrate material is significantly improved. Also, in the case of the adhesive tape that has been manufactured according to the present invention, the cohesive strength of the adhesive agent itself is significantly increased and because of that a material is obtained where the durability properties, and the thermal resistance properties are improved and not only that but also it is a material that has excellent holding properties.

Also, regarding the adhesion between the adhesive agent and the tape substrate material, it becomes extremely strong and not only that but also, the phenomenon where with the passing of the time a separation between the adhesive agent and the tape substrate material occurs, is not observed at all, and even in the case when it is stored for a long period of time there is no plasticising of the adhesive agent and leakage from the side surface and deterioration of the appearance.

Practical Example 1

100 parts of SBR (JIS-11502 manufactured by Nippon Gosei Rubber Company) was kneaded by using a double roll and after that it was dissolved in 50.0 parts of cyclohexanone. To that, as the adhesion imparting agent, 50 parts of Pikkopel (manufactured by Pennsylvania Industrial Chemical Company) were added, and as the plasticising agent, 10 parts of dioctyl phthalate (manufactured by Sekisui Chemical Industries Company) were added, and as the anti-ageing agent, 2 parts of Antiage (manufactured by Kawako Chemical Company) were added, and after that especially, methyl

methacrylate was added so that the concentration of the methyl methacrylate monomer material was made to be 30 %.

This material was coated on the surface of 0.15 mm thick polyethylene film (Sumikasen F205, manufactured by Sumitomo Chemical Company), and by using a resonant transformer type electron accelerating device (E.B.G., manufactured by General Electronics Company) a 2MeV electron beam was irradiated and after that this was dried by using warm air flow at a 50oC for a period of 30 minutes, and especially, it was stored overnight and after that its tape performance was measured.

The results from these measurements are shown in Table 1.

Table 1

第 1 表

	単位	1	2	3	4	5
照射線量	Mrad	0	3	5	7	10
粘着力	1／32″	10	9	8	8	7
接着力	g／19㎜	糊面剝離	500	700	610	550
投錨力	g／19㎜	400	450	670	950	1500以上

In the table:

1. radiation beam amount, 2. adhesive strength, 3. bonding strength, 4. anchoring strength, 5. units, 6. above.

Practical Example 2

100 parts of a two-element copolymer material formed from 90 parts of butyl acrylate and 10 parts of methyl methacrylate, were dissolved in 370 parts of benzene. To that, as the vinyl monomer material, acrylamide ...parts (there is no number – translator's note) and 10 parts of butyl acrylate, were added, and especially, by using a catalyst a preliminary polymerization was conducted until the viscosity became approximately 4000 CP (polymerization coefficient of approximately 20 %). This prepolymerized material was then coated on the surface of 0.15 mm thick

polyethylene film, according to the same method as described in the Practical Example 1, and by the irradiation using an electron beam, the adhesive tape was produced. The results from the measurements of the tape performance at this time are shown according to the presented in Table 2.

Table 2

	単位	6	7	8
照射線量	Mrad	0	3	7
粘着力	1/32"	11	11	11
接着力	g/19㎜	520	550	600
投錨力	g/19㎜	845	990	1230

In the table:

2. radiation beam amount, 2. adhesive strength, 3. bonding strength, 4. anchoring strength, 5. units.

In term "parts" in the above described practical examples means "weight parts" in all cases. Also, the measurements were conducted according to the described here below methods.

Adhesive Strength Measurement Method

This measurement is done according to the ball test method. This is done as on a 30o tilted surface the adhesive tape is fixed so that its adhesive surface is on the top, and 32 steel balls with a diameter obtained as a 1 inch diameter is divided into 32 equal parts, are slipping on a suitable 10 cm non-adhesive surface and then after that the measurement results is indicated by the number of the largest diameter of the ball that has stopped on the adhesive surface of the subsequent 10 cm adhesive tape.

Bonding Strength Measurement Method

The measurement is conducted by the peel test method. The adhesive tape is glued on a stainless steel plate and a 2 kg rubber roll is passed back and forth twice and after that the adhesive tape is peeled back relative to the test panel at an angle of 180 degrees, and the separation strength is measured at the time when there is separation, using a speed of 300 mm/min.

Anchoring Strength Measurement Method

Vinyl adhesive tape is glued onto the experimental tested adhesive tape and then a 2 kg rubber roller is passed back and forth twice, and after that at a speed of 300 mm/min, a separation experiment is conducted so that it is peeled back at an angle of 180 degrees, and the separation strength is measured at the time when the adhesive agent of the experimental tape was separated from the tape substrate material.

(57) Scope of the Claims of the Invention

1. Manufacturing method for the preparation of a pressure sensitive adhesive tape characterized by the fact that an adhesive agent, containing vinyl type monomer material, is provided on the surface of a tape substrate material formed from synthetic resin film, and then this is irradiated with ionization irradiation beam.

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⑤4感圧性接着テープの製造方法

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発明の詳細な説明

本発明はテープ基材から粘着剤が剝離することのない秀れた粘着テープを製造する方法に関するものである。特に本発明はポリエチレン、ポリプロピレン、テトラフルオロエチレン等の比較的接着性に乏しい合成樹脂フイルムを基材とした粘着テープを製造するに適した方法に関するものである。

従来、ポリエチレン、ポリプロピレン、テトラフルオロエチレン等の合成樹脂をテープ基材として用いた場合には、該合成樹脂フイルムが化学的、電気的に極めて秀れた特性をもつものではあつたが、粘着剤との接着性が悪いので良好な粘着テープを製造することが困難であつた。しかしながら、これまでに接着性に乏しい合成樹脂フイルムと粘着剤の接着を強固にする為に種々の研究がなされている。中でも、特公昭40—24788号公報に記載せられる如く、還元性の増感剤を混入せしめた粘着剤をテープ基材面に塗布した後、テープ背面より可視及び紫外線を照射することによつて、テープ基材と粘着剤との間の接着を強化できることが知られている。しかし乍ら、この様に可視及び紫外線を照射することによつて、テープ基材と粘着剤との境界面で両者間に架橋反応をおこし、もつて投錨力を増大せしめる方法に於いては、テープ基材としては比較的厚味が薄く、しかも透明性の良好なものに限定される欠点があり、又この様にして得られた粘着テープはたとえ照射線に対する増感剤を粘着剤中に配合させて連続的に架橋化反応を行つたとしても、かゝる架橋化反応を完全に終結させることが困難なので、日が経過するにつれて序々に架橋化反応が進行して粘着性を損う傾向が強いのである。

更に又、この方法によつてもポリエチレン、ポリプロピレン、テトラフルオロエチレン等の樹脂をテープ基材としたときにはテープ基材と粘着剤との接着力を充分にすることはできなかつた。

しかして、本発明は上述の如く反応を調節することが極めて困難な架橋反応を利用するものではなく、電離性放射線を照射することによつて粘着剤中に加えたビニール系単量体を重合すると同時にテープ基材並びに粘着剤成分へも一部グラフト重合せしめることによつて、粘着剤の基材への投錨力を極めて向上させたものである。

即ち、本発明はビニール系単量体を含有せる粘着剤を、合成樹脂フイルムよりなるテープ基材面に設け、さらにこれに電離性放射線を照射することを特徴とする感圧性接着テープの製造方法に関するものである。

本発明に用いられる粘着剤とは、一般に用いられる粘着テープ用糊剤をすべて総称するものである。一般に用いる粘着剤はゴム或は合成樹脂に粘着賦与剤を加えてなり、更に必要に応じて軟化剤、酸化防止剤、充填剤、顔料等を混入したものであるが勿論これらはすべて本発明に利用できる。或いは又ビニール系単量体を適宜合成することによつて得られた重合物であつてもよい。

ゴムとしては天然ゴム、スチレンーブタジエンゴム、ニトリルゴム、ブチルゴム、ネオプレンゴム、合成天然ゴム等が使用され、又合成樹脂としては塩化ビニール樹脂、酢酸ビニール樹脂、塩ビー酢

ビ共重合体等の熱可塑性樹脂及びビニール系単量体等を適宜合成して得られる重縮合物が用いられる。

又粘着賦与剤としてはαおよびβピネンを主成分とするテルペン系樹脂、アビエチン酸を主成分とするロジン系樹脂及びロジン系樹脂に水素添加したり、エステル化したりして得られるロジン変性体、オレフイン類より得られる炭化水素系樹脂等が使用される。

本発明に用いられる粘着剤組成物としてはかゝるゴム系或いは合成樹脂系粘着剤にビニール系単量体を混入することによつて得られるのである。

ビニール系単量体としては常温に於いて気状、固状のいずれでも使用できるが、スチレン及びスチレン誘導体、メタクリル酸、アクリル酸、アクリル酸アミド、酢酸ビニール、マレイン酸エステルの如き不飽和二塩基酸誘導体、アクリル酸エステル、メタクリル酸エステル、ビニルエステルおよびビニルエーテル等通常室温にて液状のもの或は固状のものが粘着剤への混入操作を容易にするので好適である。

特にアクリル酸ブチル、酢酸ビニール、メチルメタアクリレートが好ましい。

テープ基材としては合成樹脂フイルムであれば如何なるものでも効果があり、特に限定しないが、従来の方法では基材と粘着剤との間の接着が困難であつたポリエチレン、ポリプロピレン、テトラフルオロエチレン等の合成樹脂のフイルムをテープ基材としたときに特に効果的である。次に本発明による粘着テープの製造方法について、その概要を述べる。

まず、粘着剤の主成分となる天然ゴム或いは合成ゴムをロール又はバンバリーミキサーによつて素練りした後適当に切断して、予め有機溶剤を入れた溶解タンクに投入する。更にこれに適当量の粘着賦与剤、及びビニール系単量体を又必要に応じて軟化剤や老化防止剤を順次混入し、加熱しながら攪拌溶解する。かくして得られた粘着剤組成物をロール・コーター、ナイフコーター等のコーター等のコーターによつてテープ基材の片面もしくは両面に厚味が一定になるように塗布する。続いてこのテープの片面より電離性放射線を照射せしめて後、乾燥機を通過させるのである。粘着剤中の溶剤及び未反応のビニール系単量体は乾燥機を通過する間にほゞ完全に揮散されてしまうのである。

尚、上記方法は粘着剤組成物の溶液を塗布し、電離性放射線を照射後乾燥させたが、粘着剤組成物の溶液を塗布し乾燥して粘着剤中の溶剤及び余分のビニール系単量体を揮散せしめた後に電離性放射線を照射させても同様な結果が得られる。

更に本発明に於いては次の如き変形も可能である。即ち天然ゴム或いは合成ゴムを二本ロール又はバンバリーミキサーによつて素練りすると共に、これに適当量の粘着賦与剤、軟化剤、充填剤、老化防止剤及びビニール系単量体を順次混入混練して固形状の粘着剤組成物を得る。

かくして得られた粘着剤組成物をカレンダー等によつてテープ基材の片面上に厚味が一定になるように塗布する。続いてこのテープの片面より電離性放射線を照射せしめて後、これをシート状のまゝ一定の長さに切断するか或いは適宜の長さに巻取つて後一定の長さに切断してロール状の製品とするのである。

更に本発明に於いてはビニル系単量体を電離性放射線を照射して重合を行う前に触媒により一部重合させて、その後電離性放射線で重合しても良い。

本発明によればテープ基材面に、ビニール系単量体を含む粘着剤組成物を塗布し、これに電離性放射線を照射することにより粘着剤中に含有されたビニール系単量体それ自体が重合すると同時に、テープ基材面並びに粘着剤成分にもビニール系単量体の一部がグラフト重合するものと考えられ、テープ基材への投錨力は著しく改善されるのである。又本発明によつて製造された粘着テープは、粘着剤自体の凝集力が著しく向上するので耐老化性、耐熱性が改良されるばかりではなく、すぐれた保持力を有するものである。

又粘着剤とテープ基材との接着は極めて強固となりしかも経日によつて粘着剤とテープ基材が剝離する現象は全く認められず、長期保存後に於いても粘着剤が軟化して側面からしみ出して外観を損うことがないのである。

実施例　1

ＳＢＲ（ＪＩＳ－１５０２日本合成ゴム社製）１００部を二本ロールで素練りした後５００部のシクロヘキサンに溶解させる。これに粘着附与剤

5

としてピッコペール（ペンシルバニア・インダストリアル・ケミカル社製）５０部、軟化剤としてジオクチルフタレート（積水化学工業社製）を１０部、老化防止剤としてアンテイゲ（川口化学社製）２部を加えて後、更にメチルメタクリレートを加えてメチルメタクリレート単量体の濃度を３０％とした。

これを厚味0.１５㎜のポリエチレンフイルム※（スミカセンＦ２０５住友化学社製）面上に塗布し、レゾナント、トランスフオーマー型電子加速器（ゼネラルエレクトリック社製Ｅ・Ｂ・Ｇ）により２MeV の電子線を照射した後、これを５０℃で３０分間熱風により乾燥せしめ更に一昼夜放置した後テープ性能を測定した。

その結果を第１表に示す。

第　1　表

	単位	1	2	3	4	5
照射線量	Mrad	0	3	5	7	１０
粘着力	1／３２″	１０	９	８	８	７
接着力	g／１９㎜	糊面剝離	５００	７００	６１０	５５０
投錨力	g／１９㎜	４００	４５０	６７０	９５０	１５００以上

実施例　2

アクリル酸ブチル９０部とメチルメタアクリレート１０部とから得られた二元共重合体１００部を３７０部のベンゼンに溶解させる。これにビニル単量体としてアクリルアマイド部とアクリル酸ブチル１０部を加え、更に触媒により粘度が約４０００CP（重合率約２０％）となるまで予備重合を行つた。此の予備重合物を実施例１と同様な方法で0.１５㎜のポリエチレンフイルム面上に塗布し、電子線を照射することによつて粘着テープを作成した。その際のテープ性能の測定結果を第２表に示す。

第　2　表

	単位	6	7	8
照射線量	Mrad	0	3	7
粘着力	1/32″	11	11	11
接着力	g/19㎜	520	550	600
投錨力	g/19㎜	845	990	1230

但し上記実施例中部とあるのはすべて重量部を表す。又測定方法は次の方法により行つた。

粘着力の測定方法

ボール・テスト法に依る。これは３０° の斜面に糊面が上面になるようにして粘着テープを固着し、１インチの径を３２等分した３２個の鋼球を適宜１０㎝の非粘着面を助走させてころがし、それに続く１０㎝間の粘着テープ糊面上で止まる最大径番号で示す。

接着力測定方法

ピール・テスト法に依る。ステンレス・スチール板に粘着テープを貼着し、2kgのゴムロールを２往復させた後、粘着テープをテストパネルに対して１８０° に折り返して、３００㎜／min の速度で剝離したときの剝離力を測定した。

投錨力の測定方法

ビニール粘着テープを試料粘着テープに貼り合せ2kgのゴムローラーを２往復させた後、３００㎜／min の速度で試料面に対し１８０° になるようにして剝離試験を行い、試料テープの粘着剤がテープ基材より剝離したときの剝離力を測定した。

㊲特許請求の範囲

1　ビニール系単量体を含有せる粘着剤を合成樹脂フイルムよりなるテープ基材面に設け、さらにこれに電離性放射線を照射することを特徴とする感圧性接着テープの製造方法。

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